

AMENDMENT UNDER 37 C.F.R. § 1.114(c)  
Application No.: 09/732,712  
Atty Docket No.: Q57601

### **REMARKS**

In response to the Office Action of March 24, 2004, applicants have filed a Request for Continued Examination (RCE) in order to gain entry of the amendments that are set forth above, which are based on various telephone conversations that applicants' undersigned counsel had with the Examiner, Ms. Gakh. These telephone conversations are summarized below.

Claims 1 to 10, 13 to 16 and 22 to 24 are all the claims pending in the application, prior to the present amendment.

In the Office Action of March 24, 2004, the Examiner sets forth objections to the specification and various rejections of the claims in Paragraphs 3, 5, 8 11, 12 and 13 of the Office Action. These objections and rejections are identical to the objections and rejections in Paragraphs 3, 4, 6, 9, 10 and 11, respectively, of the previous Office Action of July 18, 2003.

In addition, in Paragraph 6, of the present Office Action, the Examiner sets forth a new rejection of the newly added claims 23 and 24.

Further, in Paragraph 14 of the present Office Action, the Examiner responds to the various remarks and arguments that applicants submitted in the Amendment of January 20, 2004.

On June 30, 2004, applicants' undersigned counsel called the Examiner, Ms. Gakh, to schedule a personal interview. Applicants believed based on the various comments that the Examiner set forth in the Office Action that there was a misunderstanding of the present invention by the Examiner.

Applicants' undersigned counsel then had telephone discussions with the Examiner on June 30, 2004, the following day July 1, 2004, and then again on July 7, 2004. As a result of

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those discussions, applicants have amended the claims as set forth above. Applicants believe that the above amendments overcome all of the objections and rejections that are in the application.

The following comments summarize the various telephone conversations the undersigned counsel had with the Examiner.

As a general matter, applicants' undersigned counsel first pointed out that the prior art cited by the Examiner to reject the claims does not disclose or suggest using a gaseous phase moiety of liquefied ammonia as a reference gas, and, therefore, applicants did not understand why the claims were rejected over the cited prior art. The Examiner agreed that the cited prior art did not disclose or suggest using a gaseous phase moiety of liquefied ammonia as a reference gas. The Examiner stated that her main problems with the allowability of the claims were based on enablement and indefiniteness.

Accordingly, the Examiner agreed that the present application contains allowable subject matter for those embodiments of the invention for which there is sufficient enablement.

Applicants' undersigned counsel then discussed with the Examiner questions of enablement and indefiniteness. The Examiner proposed various amendments to overcome the problems. Applicants have now amended the claims in a manner that was agreed to by the Examiner to address the issues which the Examiner stated existed with respect to enablement and indefiniteness, except for certain amendments in the last two steps of claim 1 that applicants will discuss below which differ from the amendments proposed by the Examiner. Applicants believe that the amendments that they have made in the last two steps accurately define the invention.

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Applicants' undersigned counsel notes that not all of the specific issues that are set forth in the Office Action were discussed with the Examiner, but the Examiner indicated that the proposed amendments addressed all of the issue with which she was concerned.

Turning now to the specific amendments proposed by the Examiner, the Examiner recognized that the specification discloses that the sample which is measured can be either ammonia gas or liquefied ammonia, but took the position that the specification provides enablement only for the use of liquefied ammonia as the sample. Accordingly, the Examiner proposed amendments to claim 1 to direct claim 1 to measuring the water concentration in liquefied ammonia.

Thus, the Examiner proposed to amend the preamble to claim 1 to state that the sample being measured was "liquefied" ammonia. The Examiner also proposed to amend the preamble to state that the sample has a water concentration of no more than 10 ppm, and to cancel claim 6 which recited this concentration. Applicants have amended the preamble in the manner agreed upon with the Examiner, and have canceled claim 6 as proposed by the Examiner.

For ease of discussion, applicants refer to the six subparagraphs of claim 1 as paragraphs (a) to (f), respectively, and discuss these paragraphs below.

(a) The Examiner indicated that she wished to amend paragraph (a) to make it clear that the water concentration in the reference gas in paragraph (a) did not refer to the water concentration in the gaseous phase moiety, but to the water concentration in the liquefied ammonia from which the gaseous phase moiety was taken. Applicants have amended this paragraph in the manner agreed upon with the Examiner.

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(b) The Examiner stated that what was being measured by the spectrometer was the infrared spectrum of the reference gas, and not the absorption intensity of the reference gas.

The Examiner relied on the disclosure at page 13, lines 1 to 5 of the specification for the language of step (b). This portion of the specification describes measurements made with the reference gas and states that “the infrared absorption spectrum is measured” and that the “results obtained are used as the background of the infrared spectroscope”.

Applicants have amended this step (b) in the manner agreed upon with the Examiner.

(c) The Examiner stated that the specification provides enablement only for the embodiment of the invention where the sample is liquefied ammonia, and that this embodiment requires that the liquefied ammonia sample be vaporized. Step (c) has been amended to follow the language in claim 5 (which has been canceled) and the disclosure at page 13, lines 11 to 15, in the manner agreed upon with the Examiner.

(d) The Examiner stated that what was being done in this step was the measuring of the IR spectrum of the sample employing the background of the spectrometer. The Examiner stated that Figure 4, which is discussed in the last paragraph of page 13 of the specification, shows that IR spectrum is being measured. Accordingly, step (d) has been amended to specifically refer to the IR spectrum of the sample and the background of the spectrometer, in the manner agreed upon with the Examiner.

(e) Step (e) did not appear in the previous version of claim 1, and therefore is an entirely new step.

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The Examiner stated that her understanding of how the process works is that after the two IR spectrums are measured, then the absorption intensity at an infrared wave number is measured. The Examiner also stated that her understanding was that what is measured is “water absorption intensity”. Based on these understandings, the Examiner proposed a step (e) to set forth the step that applicants were performing.

Applicants in claim 1 as amended above have set forth a step (e) that is similar to, but different from the step (e) proposed by the Examiner. In the following paragraph, applicants have indicated applicants’ changes from the Examiner’s proposal by underlining to show additions and brackets to show deletions.

--(e) measuring [water] absorption intensity in the IR spectrum of the sample at an infrared wave number at which water absorbs IR and at which infrared absorptions of ammonia and water do not overlap, and--

Applicants submit that step (e) as set forth in claim 1 as amended above accurately sets forth the step that is being employed in the present invention.

(f) In step (f), the Examiner proposed using the word “determining” based on the description at page 10, lines 5 to 13 where the words “determination” and “determining” were used. Applicants have amended step (f) to employ the word “determining” as proposed by the Examiner.

Further, the Examiner proposed amending step (f) to refer to the “measured water absorption intensity” described in her proposed step (e). Applicants have deleted the term “water” from this phrase in view of the manner in which applicants have amended step (e).

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In addition, applicants have employed the term --from-- instead of the word “using” that the Examiner proposed.

Applicants submit that step (f) as set forth in claim 1 as amended above accurately sets forth the step that is being employed in the present invention.

Turning now to the remaining claims, the preamble has been amended in claims 2 to 4 and 7 to 10 by adding the word “liquefied” to make it consistent with the preamble of claim 1, in the manner agreed upon with the Examiner.

In addition, in each of the claims 2 to 4, the word “infrared” has been inserted and the words “measurement” and “used” have been deleted, for better consistency with amended claim 1, in the manner agreed upon with the Examiner.

With respect to claim 3, the Examiner raised new questions with respect to the phrase “one or more”. The Examiner stated that she did not understand how more than one wave number could be used for the measurement.

Further, the Examiner questioned whether water has infrared absorption at each of the wave numbers recited in claim 3. The Examiner referred applicants’ undersigned counsel to the following website <http://www.lsbu.ac.uk/water/vibrat.html> which discusses the absorption of water. Applicants attach a copy of pages 1 to 4 of a printout from this website. The Examiner particularly referred to the table at page 1 of the enclosed printout, and referred to the absorptions of 3657.05 and 3755.93  $\text{cm}^{-1}$  for  $\nu_1$  and  $\nu_3$ , respectively. The Examiner stated that she could, therefore, understand the 3656 and 3756 numbers in claim 3, but was surprised that water absorptions occur at the other wave numbers recited in claim 3.

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In response, although it is possible to measure water concentration using one wave number, a measurement method where two or more wave numbers are used and the average value is obtained from the results is more reproducible.

The basic molecular vibration modes in the water molecules are symmetric stretching vibration, bending vibration and asymmetric stretching vibration. In addition to the molecular vibration, the molecule rotates, so that the rotational energy levels combine with each of the vibrational energy levels. In a case like the present invention where water content in a gas is measured, with the molecules freely rotating, the transition occurs not only between the vibrational levels but between rotational levels in each of the vibrational levels, which results in that water absorptions occur at many wave numbers.

Accordingly, the absorptions for water are not limited to the  $\nu_1$  and  $\nu_3$  absorptions disclosed in the table.

In view of the above, applicants submit that the specification enables one of ordinary skill in the art to practice the invention of claim 3.

With respect to claims 5 and 6, the recitations of these claims, in effect, have been incorporated into claim 1. Therefore, claims 5 and 6 have been canceled.

With respect to claim 7, the dependency of claim 7 has been changed to claim 1, in view of the cancellation of claim 6.

With respect to claims 22 and 24, these claims were specifically directed to measuring water concentration in liquefied ammonia and have now been canceled, since claim 1 is now directed to such a method.

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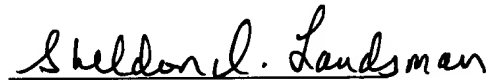
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In view of the above, applicants believe that the above amendments overcome all of the objections and rejections, and request their withdrawal.

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,



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WASHINGTON OFFICE

**23373**

CUSTOMER NUMBER

Date: July 26, 2004





## Molecular Vibration and Absorption

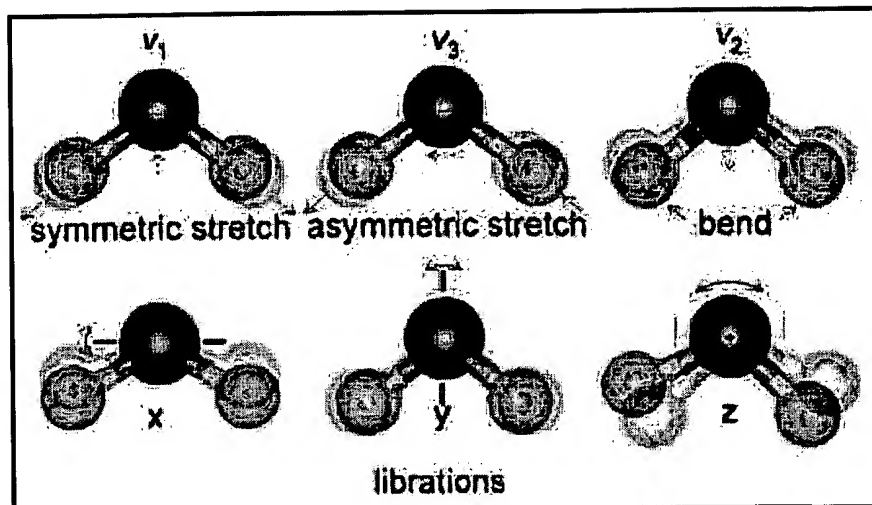
Water is the main absorber of the sunlight. The 13000 billion tons of water in the atmosphere removes about 70% of the radiation, mainly in the infrared region where water shows strong absorption. It contributes significantly to the greenhouse effect ensuring a habitable planet, but operates a negative feedback effect, due to cloud formation, to attenuate global warming.

Water vapor spectroscopy has been recently reviewed [348]. The **water molecule** may vibrate in a number of ways. In the gas state, **the vibrations** [607] involve combinations of symmetric stretch ( $\nu_1$ ), asymmetric stretch ( $\nu_3$ ) and bending ( $\nu_2$ ) of the covalent bonds with absorption intensity ( $\text{H}_2^{16}\text{O}$ )  $\nu_1; \nu_2; \nu_3 = 0.07; 1.47; 1.00$  [8].

	$\nu_1, \text{cm}^{-1}$	$\nu_2, \text{cm}^{-1}$	$\nu_3, \text{cm}^{-1}$
$\text{H}_2^{16}\text{O}$	3657.05	1594.75	3755.93
$\text{H}_2^{17}\text{O}$	3653.15	1591.32	3748.32
$\text{H}_2^{18}\text{O}$	3649.69	1588.26	3741.57
$\text{HD}^{16}\text{O}$	2723.68	1403.48	3707.47
$\text{D}_2^{16}\text{O}$	2669.40	1178.38	2787.92
$\text{T}_2^{16}\text{O}$	2233.9	995.37	2366.61

In liquid water and ice the infrared and Raman spectra are far more complex due to vibrational overtones and combinations with librations (restricted rotations; *i.e.* rocking motions) due to the hydrogen bonds (minor  $L_1$  band  $395.5 \text{ cm}^{-1}$ ; major  $L_2$  band  $686.3 \text{ cm}^{-1}$ ; both at  $0^\circ\text{C}$ , the absorbance of  $L_1$  increasing with temperature whereas  $L_2$  absorbance decreases but broadens with reduced wavenumber with temperature [177]), intermolecular stretching (S band  $183.4 \text{ cm}^{-1}$ ; at  $0^\circ\text{C}$ , the absorbance increasing with temperature to about  $30^\circ\text{C}$  before reducing with temperature due to hydrogen bond breakage [608]) and bending (B band  $\sim 40 - 60 \text{ cm}^{-1}$ ; at  $0^\circ\text{C}$ , the absorbance increasing with temperature [608]) of the hydrogen bonds, and cluster vibrations such as translational vibrations involving combinations of stretching and bending at around  $200 \text{ cm}^{-1}$  [240] ( $229 \text{ cm}^{-1}$  in ice 1h). Also in liquid water there is a small but significant combination band (IR and Raman) of the bending ( $\nu_2$ ) and libration modes at  $2125 \text{ cm}^{-1}$  ( $2270 \text{ cm}^{-1}$  in ice 1h; the 'association band'). Variations in the environment around each liquid water molecule gives rise to considerable line broadening. A contribution to the Raman scattering of water at  $50 \text{ cm}^{-1}$  has been attributed to the low-frequency vibrations of oxygen-oxygen bonds at the vertices of pentagonal dodecahedral structures [165]. Such low-frequency features (centered around  $60 \text{ cm}^{-1}$ ), however, are also observed in the spectra of dense non-associated liquids, such as the noble gasses, and are best attributed to non-bonded 'cage effects' where translations are frustrated [448]. The spectra for isotopic variants of water (*e.g.* HDO,  $\text{D}_2\text{O}$  and  $\text{H}_2^{18}\text{O}$ ) are all different; in particular the H-O and D-O stretching vibrations are not connected in HDO but the related vibrations in  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  involve both hydrogen atoms.

Shown opposite are the main vibrations occurring in water. The movements are animated on **another page**. The dipole moments change in the direction of the movement of the oxygen atoms as shown by the arrows.



The main stretching band in liquid water is shifted to a lower frequency ( $\nu_3$ , 3490  $\text{cm}^{-1}$  and  $\nu_1$ , 3280  $\text{cm}^{-1}$  [8]) and the bending frequency increased ( $\nu_1$ , 1645  $\text{cm}^{-1}$  [8]) by hydrogen bonding; stretching frequency increasing about 360 (at 3.1 Å) -1000 (at 2.6 Å)  $\text{cm}^{-1} \text{Å}^{-1}$  with increasing O...O distance and about 9  $\text{cm}^{-1} \text{degree}^{-1}$  with increasing O-H...O bend [446]. The absorption intensity of these bands is  $\nu_1;\nu_2;\nu_3 = 0.87;0.33;1.00$  [8]. In ice, these bands are shifted further ( $\nu_1$ , 3085  $\text{cm}^{-1}$ ,  $\nu_2$ , 1650  $\text{cm}^{-1}$ ,  $\nu_3$ , 3220  $\text{cm}^{-1}$ ). In liquid water the molecular stretch vibrations shift to higher frequency, on raising the temperature or pressure (as hydrogen bonding weakens the covalent O-H bonds strengthen, causing them to vibrate at higher frequencies) whereas the intramolecular and molecular bend vibrations shift to lower frequencies. Raising the temperature also lowers the intensity of the stretching bands. This divergent behavior of bending and stretching vibrations allows their contributions to combination bands to be discerned. Thus, combinations of stretching vibrations shift to higher frequency with temperature with this trend reduced when bending vibrations are also combined. As examples, the first overtone combination of symmetric and asymmetric stretching shows a shift from strongly hydrogen-bonded structures (6707  $\text{cm}^{-1}$ ) to weakly hydrogen bonded structures (7082  $\text{cm}^{-1}$ ) with increasing temperature [237], and the combination band at about 5200  $\text{cm}^{-1}$  shifts to slightly higher wavenumbers with reduced hydrogen bond strength [282]. The second overtone of the stretching band gives rise to a significant peak in the near-infrared spectrum ( $\lambda$  970 nm). Recent work shows that the intensity of all overtone bands tend to increase linearly with increasing temperature in the range 2°C - 85°C without isobestic points (in contrast to earlier reports [530]), so ruling out a simple two-phase model for water [526] but more supportive of progressively changing two-phase models such as **described here**.

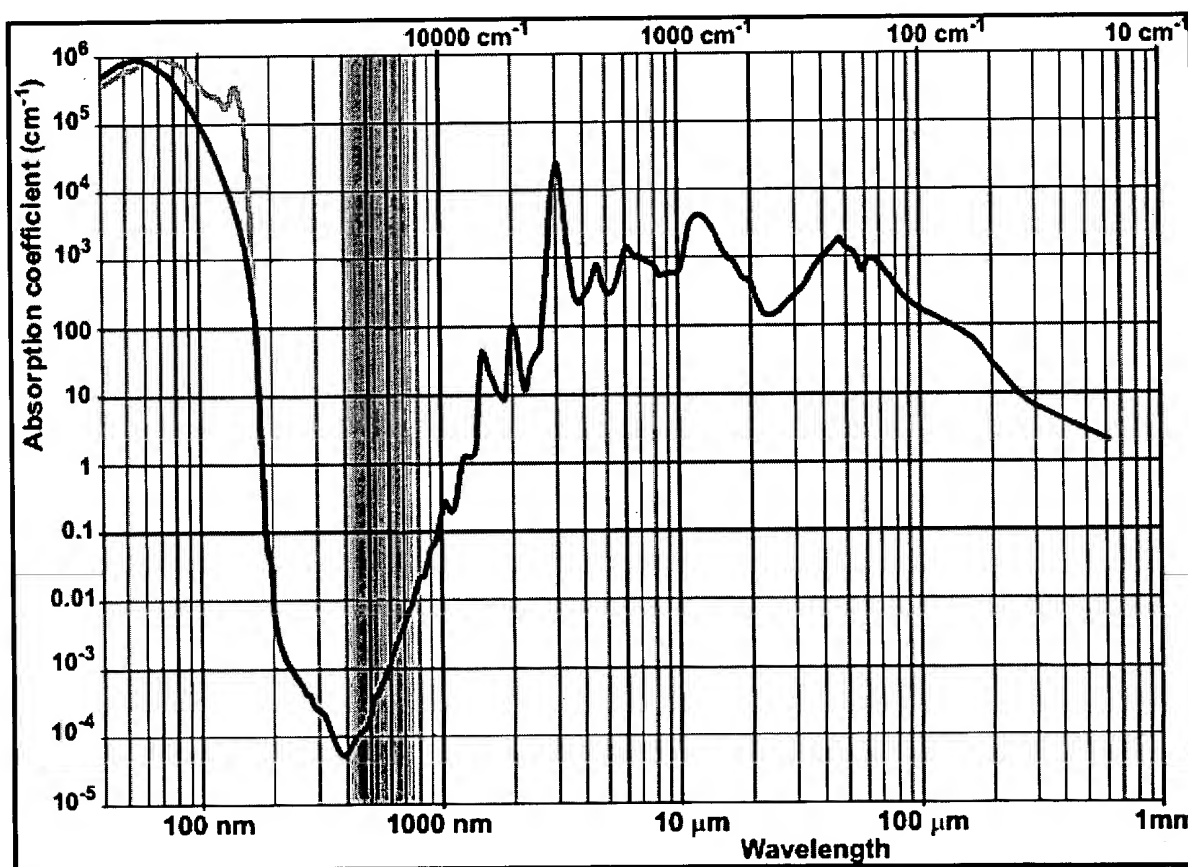
**Assignment of the vibrational absorption spectrum of liquid water**

Wavelength	$\text{cm}^{-1}$	Assignment	Wavelength <sup>a</sup>	$\text{cm}^{-1}$	Assignment
0.2 mm	50	intermolecular bend	1470 nm	6800	$a\nu_1 + b\nu_3; a+b=2$
55 $\mu\text{m}$	183.4	intermolecular stretch	1200 nm	8330	$a\nu_1 + \nu_2 + b\nu_3; a+b=2$
25 $\mu\text{m}$	395.5	$L_1$ , librations	970 nm	10310	$a\nu_1 + b\nu_3; a+b=3$
15 $\mu\text{m}$	686.3	$L_2$ , librations	836 nm	11960	$a\nu_1 + \nu_2 + b\nu_3; a+b=3$
6.08 $\mu\text{m}$	1645	$\nu_2$ , bend	739 nm	13530	$a\nu_1 + b\nu_3; a+b=4$

4.65 $\mu\text{m}$	2150	$\nu_2 + L_2$	660 nm	15150	$a\nu_1 + \nu_2 + b\nu_3$ ; $a+b=4$
3.05 $\mu\text{m}$	3277	$\nu_3$ , asymmetric stretch	606 nm	16500	$a\nu_1 + b\nu_3$ ; $a+b=5$ [526]
2.87 $\mu\text{m}$	3490	$\nu_1$ , symmetric stretch	514 nm	19460	$a\nu_1 + b\nu_3$ ; $a+b=6$ [526]
1900 nm	5260	$a\nu_1 + \nu_2 + b\nu_3$ ; $a+b=1$	Note that a and b are integers, $\geq 0$		

<sup>a</sup> Wavelength (nm) =  $10^7/\text{wavenumber}(\text{cm}^{-1})$   $\sim 3.3$  attosecond

The near-infrared (NIR) bands (at about  $\lambda$  970-1940 nm) are suited to rapid non-destructive water determination [479], all shifting a few nm to longer wavelength (lower frequency) with strengthening hydrogen bonding due to shifts from high density water (*i.e.* increasing CS) to low density water (*i.e.* increasing ES) [489]. A shoulder at about  $3250\text{ cm}^{-1}$  on the side of the only strongly active Raman peak (symmetric O-H stretch,  $\nu_1$ ) of liquid water has been assigned to the collective in-phase symmetric O-H vibrations of strongly tetrahedrally-bonded water patches. The ratio of this to the remaining peak at about  $3400\text{ cm}^{-1}$  has been used to determine the fraction of such water but such comparisons, although commonly used, should be treated with caution, as their absorbances are unlikely to be identical. There is clearly much structural information hidden in the vibrational spectra of water, if only it can be unambiguously interpreted. Some success has recently been made using femtosecond mid-infrared nonlinear spectroscopy [189, 190] and the theoretical Raman spectra of water clusters [483].



The spectrum of liquid water

Water is blue [131] as overtone and combination vibrational absorption bands (albeit far less intense, see above [130]) extend through the red part of the visible spectrum with a small peak at 739 nm and

shoulder at 836 nm, both varying somewhat with temperature [268] plus a smaller fourth overtone of the  $\nu_1:\nu_3$  stretch at 606 nm, and very small fifth overtone (at 514 nm) and combined overtone (at 660 nm) bands. This absorption spectrum of water (red light absorbs 100 times more than blue light), together with the five-times greater scattering of blue light over red light, contributes to the blue color of lake, river and ocean waters. Colloidal silica may contribute to the outstanding blue color of certain, often hydrothermal, pools and lakes [372]. Ice is also blue [159] for similar reasons but liquid  $D_2O$  does not absorb in the red region (as the absorption band is shifted into the infrared) and is blue solely because of the light scattering effect [159].

The first peak in the far UV of gaseous water ( $\sim 140$  nm) is due to excitation from the unoccupied  $p_z^2$ -type molecular **1b<sub>1</sub> orbital** (interactive orbitals are available (COW only [**Plug-in, ActiveX**])). Absorption of UV close-by ( $\sim 125$  nm), excites the **3a<sub>1</sub> orbital** leading to dissociation into OH + H

(photodissociation). Such dissociation can also be achieved by consecutive absorption of two 266 nm photons [589]. Inelastic x-ray scattering studies find this far UV peak to be absent in liquid water [355], where the major peak is at about 56 nm. The infrared spectrum runs into microwave radiation at longer wavelengths. This interacts with the water dipole, moving the molecules backwards and forwards and so stretching and bending the hydrogen bonds, which generates heat. If the radiation is at too high a frequency ( $> 1000$  GHz,  $\lambda < 0.3$  mm), the molecules do not have time to react to the magnetic field changes and no heat is generated. If the radiation is at lower frequencies ( $< 1$  GHz,  $\lambda < 30$  cm), the molecules react to the magnetic field changes but so slowly that effectively no heat is generated. Pure water is almost totally transparent to such low frequency radiation. The maximum absorption varies to higher frequencies at higher temperatures when the weaker hydrogen bonding allows a quicker response to changes in the field [136]. Microwave ovens typically use radiation at 2.450 GHz ( $\lambda$  12.24 cm). More details on the response of water to microwave radiation are **available**.

The reported structuring absorption of **sound** by water is not generally accepted.

Please submit any **comments and suggestions** you may have.

**Water: Home | Microwave effects | Water molecule | H<sub>2</sub>O spectrum | (H<sub>2</sub>O)<sub>2</sub> spectrum | (H<sub>2</sub>O)<sub>5</sub> spectrum | (H<sub>2</sub>O)<sub>8</sub> spectrum | Magnetic effects | Notes**



This page was last updated by **Martin Chaplin**  
on 7 July, 2004

selected by Science Educators  
**SCILINKS.**  
from NSTA

